Enhanced and oscillatory magnetoresistance of thin Fe(001) films

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We have studied the magnetoresistance of single-crystalline (001) Fe films prepared by sputtering techniques and covered by epitaxial MgO or Pt protective layers. The influence of the thickness of the magnetic layer as well as of the nature of the capping layer on the Fe anisotropic magnetoresistance was investigated by four-terminal probe, measurements performed with the current **J** in the [110] magnetically hard direction. We found an enhancement in the magnetoresistance value with respect to bulk and oscillations in the high field regime in MgO covered Fe/MgO(001) ultrathin films. A plausible explanation is that the enhanced magnetoresistance is due to electronic confinement effects, not present in similar Pt/Fe/MgO(001) samples. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2191092]

The physical basis for the anisotropic magnetoresistance (AMR) is that the electrical resistance of ferromagnetic layers depends on the relative angle between the magnetization vector and the direction of the sensing current. It is usually explained as a spin-orbit interaction where the magnetization of the system deforms the electron cloud, thereby changing the rate of scattering and resulting in anisotropic conduction.

Regarding AMR experiments in thin Fe films, several studies using a number of substrates and capping layers have been carried out. For example, the AMR thickness dependence of epitaxial Fe films grown on GaAs(110) and with a native oxide cover had been studied earlier by Tondra *et al.*[1] They found, for 500 nm thick films, a dependence of the AMR upon the crystalline direction. The magnetoresistance in Au/Fe/Au(001) structures with Fe thickness between 0 and 2.5 nm was studied by Yuasa *et al.*,[2] observing a granular type giant magnetoresistance associated with the discontinuous nature of the Fe layer. More recently, the magnetoresistive properties of Pd capped Fe epitaxial films in the range of thickness of 5–100 nm and grown on MgO(001) were studied by Granberg *et al.*,[3] finding a change in sign in the AMR associated with a crossover from Lorentz force to spin-orbit dominated scattering process governing the AMR.

However, especially in the atomic layer thickness limit, the influence of the interaction with the substrate and capping layer is extremely important to determine the intrinsic properties of the ferromagnetic film. In addition to this, effects in the magnetotransport properties of a physical system when its dimensions are sufficiently small are expected.[4] In this sense the MgO/Fe/MgO system is ideal, as theoretical predictions about the electronic properties of the Fe/MgO interface, lead to an Fe magnetic moment close to the case of the free Fe surface.[5] The first experiments carrying out *in situ* transport measurements in freshly deposited layers of ultrathin Fe on MgO(001) were published by Liu *et al.*,[6] finding a minimum in the resistance as a function of temperature attributed to a crossover from two-dimensional (2D) disordered to a three-dimensional (3D) metallic system. As an example of clear applications, arising from an accurate control on the epitaxy in this system, we note the promising tunnel magnetoresistance values obtained on Fe/MgO(001) based epitaxial heterostructures[7] that triggered further investigations with huge magnetoresistance results in Fe/MgO/Fe tunnel junctions.[8]

Within this scope, in order to study the influence both of the reduction of dimensionality and the nature of the capping layer, in this Letter we present results on the *ex situ* magnetotransport properties of MgO and Pt capped ultrathin Fe(001) films.

Epitaxial Fe films were sputter deposited at room temperature on single crystal MgO(001) substrates in an ultrahigh vacuum system, as described elsewhere.[9] The Fe growth was confirmed to be layer by layer which allowed us to prepare ferromagnetic Fe films exhibiting cubic magnetocrystalline anisotropy and bulk magnetic moment values for coverages down to three atomic layers. Samples were either coated with MgO or Pt (~25 Å) capping layers. In some cases, Fe films were allowed to oxidize in air to produce a stable oxide surface of about 30 Å that would act as a protective layer for the Fe underneath.

The electrical resistance was measured by a dc four-probe method while the temperature was varied from 20 to 300 K in a magnetic field up to 0.5 T. In Fig. 1 (inset) we have plotted the room-temperature resistivity ρ as a function of Fe thickness. Data could be fitted approximately by the quasiclassical-size effect Fuchs-Sondheimer equation[10] and yielded a mean free path of elastic scattering of about 510 nm. This value of scattering length is surprisingly large as would correspond to extremely clean films, where the only sources of scattering would be surface roughness and dislocations developing during the growth process.[11] The total specularity parameter is close to 74%. One is inclined to assume that most of the specular reflection occurs on the bottom Fe/substrate interface, since experiments show that both MgO/Fe and Fe/MgO interfaces are spatially different

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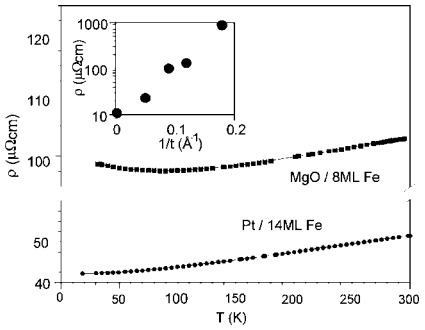

FIG. 1. Temperature dependence of the electrical resistivity measured in a magnetic field of 2500 Oe (corresponding to magnetic saturation) for an Fe thin film with MgO- (upper curve) and Pt-capping layers (lower curve). Inset: resistivity as a function of film thickness for MgO(001)-sandwiched Fe(001) ultrathin films at RT.

(the upper MgO/Fe interface being broader), with asymmetric structural defects such as dislocations and lattice distortions.[12] Figure 1 also shows a distinct temperature dependence of the electrical resistivity for two characteristic samples. For samples coated with a Pt layer, the $\rho(T)$ dependence reveals a metallic behavior, while for samples sandwiched between two MgO layers there appears a shallow minimum in the $\rho(T)$ curves, the location of the minimum gradually shifted to lower T as the film thickness was increased.[13] Such resistive minima, previously observed *in situ* by Liu *et al.*[6] in uncapped Fe/MgO layers, and related to a 2D to 3D transition, have also been studied in terms of electron-localization and/or electron-interaction effects.[4]

Figure 2(a) shows a typical in-plane magnetoresistance (MR) curve, where the measuring bias current **J** applied along the [110] magnetically hard direction and the external magnetic field **H** are both in the film plane. Here, the MR ratio is defined as $(R_H\text{-}R_{H_c})/R_{H_c}$, where R_H is the sample resistance at field H and R_{H_c} is the resistance at the coercitive field. The MR dependence on temperature (measured at a field of 2500 Oe, intense enough to assure magnetic saturation of our samples) reproduces quite well the $\rho(T)$ dependence, as is shown in the inset of Fig. 2(a). Spin-flip scattering by magnons, as well as spin-independent bulk mechanisms (phonons), may become important at higher temperatures. This indicates that resistivities due to classical size effects and spin-orbit scattering must be combined if reflections from surfaces are not completely specular.

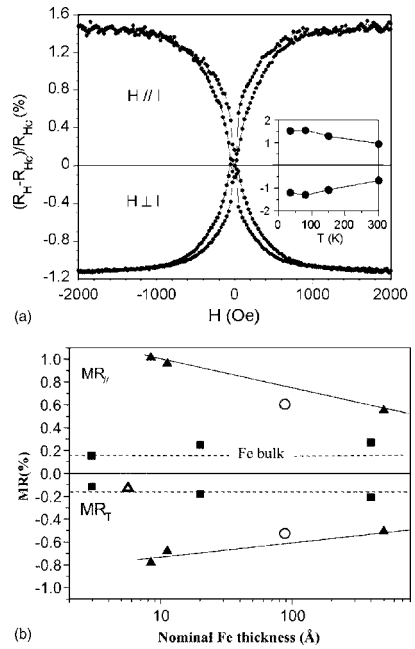

FIG. 2. (a) In-plane MR hysteresis loops of a MgO/10 ML Fe/MgO(001) sample measured at 30 K. The inset shows the change in MR as a function of temperature. (b) Anisotropic magnetoresistance as a function of Fe film thickness at RT; (■)—samples with Pt capping layer, (▲)—samples with MgO capping layers, (○)—natural oxidized Fe film, after 15 Å subtraction from nominal layer width to account for the oxide formation, and (△)—discontinuous Fe film sandwiched between MgO layers. Continuous heavy lines are guides for the eye.

In Fig. 2(b) the magnetoresistance measured in saturation is plotted as a function of Fe thickness. For samples with a Pt capping layer, the MR values are around 0.15% as for bulk Fe.[1] On the other hand, enhanced values are obtained for samples with a MgO capping layer. MR increases monotonically with decreasing Fe thickness. The similar magnetization loops for vacuum/Fe, MgO/Fe, and Pt/Fe films[9] allow us to disregard a domain wall (DW) contribution[14] to the measured magnetoresistance in MgO/Fe/MgO(001) films. In order to explore possible artifacts due to current distribution inhomogeneities, some samples were patterned in 100 μm straight line bars, with no significant modifications in the MR measurements. A comparable enhanced value is also obtained for a sample with an iron-oxide capping layer, depicted by unfilled circles in Fig. 2(b), pointing to an interpretation of this effect in terms of confinement effects. Other contributions to magnetoresistance, such as the presence of a granularlike interface alloy or Mg–O impurities in the Fe layer, would lead to an isotropic giant magnetoresistance[15] not observed in our case. Furthermore, the linearity of the current-voltage characteristics for all the films eliminates the possibility of an enhanced MR due to tunneling processes, as was also checked for a discontinuous Fe film grown on and capped with MgO layers, that presented dielectric characteristics [unfilled up triangle in Fig. 2(b)], indicating the importance of specular interfaces in order to observe enhanced MR.

In order to explain the origin of the observed phenomena, we must consider that the magnetoresistance is always reduced by spin-flip scattering. Therefore, for Fe film thicknesses close to the spin diffusion length, enhanced AMR values are expected. In the MgO/Fe/MgO(001) system, the dimensionality of electrons is reduced in the film because they cannot propagate along the [001] direction of the confinement. Besides, due to the spin-dependent reflectivity, minority electrons form quantum well states (QWSs). In particular, minority spin states cannot propagate from bcc Fe(001) into MgO(001) at E_F since there are no minority states with the same Δ_1 symmetry.[16,17] Majority states, on the other hand, are able to propagate and remain a bandlike continuum.[18] Interband scattering and the lack of specularity of the reflection due to interface roughness will tend to smear out the quantized nature of the band structure. For larger thicknesses, the three-dimensional band structure for Fe is formed and AMR tends to that of bulk Fe. For the case of Pt/Fe(001) interface there would be a strong hybridization between Fe 3*d* and Pt 5*d* states, resulting in a charge transfer that drastically decreases the minority peak[19] and thus prevents the formation of spin-polarized QWSs in the thin Fe layers.

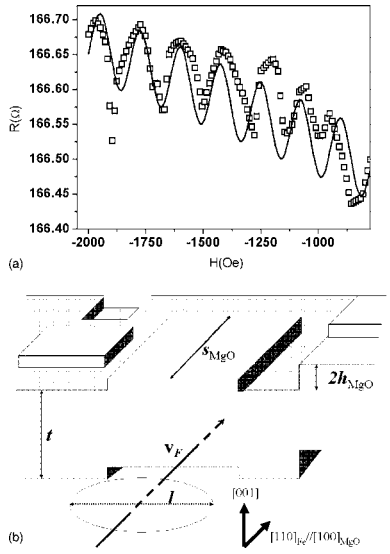

FIG. 3. (a) Resistance oscillations as a function of applied field (**H** parallel to **I**) for a MgO/10 ML Fe/MgO(001) sample. Full curve is calculated within the Altshuler-Aronov-Spivak model. (b) Schematic cross section of an Fe film of thickness t sandwiched between two MgO layers with steps of integer height $h_{MgO}=2.1$ Å and terrace length s_{MgO}. The ellipse of axis l accounts for the area with phase-coherent transport.

In addition to this, one can clearly see reproducible resistance oscillations in Fig. 3(a) with the amplitude on the order of $\Delta R/R^2 \approx 0.14e^2/h$. We estimate a period close to 180 Oe, for both polarities of the field. Similar conductance fluctuations, designated as Altshuler-Aronov-Spivak oscillations, have been previously reported in Al wires,[20] Mg cylinders and thin films,[21] as well as in 2D electron gas systems, for instance, GaAs/AlGaAs heterostructures,[22] taking as indicative of the phase-coherent transport and electron wave localization. In this case, oscillations are due to the interference of pair-electron states diffusing around closed trajectories in opposite directions with a period $\Delta B=\Phi_0/A$, where A is the area that sustains the magnetic flux. From the oscillation period in Fig. 3 and the Fe film thickness we obtain $l \approx 76$ μm as the characteristic lateral length. On the other hand, the amplitude of the oscillations may be limited by the smoothness of the interfaces, as depicted in Fig. 3(b): a conduction electron traveling with Fermi velocity close to $v_f=1.83\times10^6$ m/s [extracted from the Fermi energy of the calculated density of states[16] for each atomic layer of Fe(001) near an interface with MgO] has a de Broglie length $\lambda \approx 4$ Å that is close to the MgO(001) monolayer height, sustaining therefore scattering within the steps. As a result, oscillations in the longitudinal case are formed because the coherence transport within Fe entities extends along micrometric distances with morphology separated by MgO monoatomic heights. When the external in-plane magnetic field is applied transversal to the conductance channel, classical oscillations due to the cyclotron diameter would spread out this phase-coherence transport. Additionally, we experimentally find that the oscillations are suppressed as temperature is increased, suggesting a variation of an oscillatory density of states.

In summary, in this work we observe a nonmetallic behavior in MgO/Fe/MgO(001) samples at low temperature that is accompanied by enhanced values of AMR when compared to similar Pt/Fe/MgO(001) samples. This anomalous increase of the resistivity as well as enhanced magnetoresistance phenomena in Fe films sandwiched between oxide layers could indicate 2D confinement effects, which result from quantum interferences within the heterostructure and a distorted Fe band structure. Since this effect doubles the AMR signal at RT, it opens a new perspective on magnetic field sensing technology, especially when used with NiFe, FeCo(B), or NiCo alloys, and MgO, Ge, GaAs, or ZnSe boundaries. Oscillations in the resistivity versus H are also observed, similar to phase-coherent transport and electron wave localization effects previously observed in other systems. We believe that more work, both theoretical and experimental, is necessary on this point.

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